

# Temperature dependence of low-energy phonons in magnetic non-superconducting $\text{TbNi}_2\text{B}_2\text{C}$

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We report temperature dependence of low-energy phonons in magnetic nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  single crystals measured by inelastic neutron scattering. We observed low-temperature softening and broadening of two phonon branches, qualitatively similar to that previously reported for superconducting  $R\text{Ni}_2\text{B}_2\text{C}$  ( $R$  = rare earth, Y) compounds. This result suggests that superconductivity in  $\text{TbNi}_2\text{B}_2\text{C}$  compounds is absent not because of weak electron-phonon coupling but as a result of pair breaking due to magnetism.

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## I. INTRODUCTION

In some rare-earth nickel borocarbides  $R\text{Ni}_2\text{B}_2\text{C}$ , superconductivity coexists with magnetic order [1, 2]. Extensive neutron and x-ray scattering experiments revealed an incommensurate magnetic structure below approximately 15 K in both superconducting Er, Ho [3–6] and nonsuperconducting Tb, Gd [7, 8] compounds. This observation was interpreted in terms of common Fermi-surface nesting features along  $a^*$ , which cause magnetic ordering of the rare-earth moments via the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism [7].  $^{57}\text{Fe}$  Mössbauer spectroscopy and muon-spin relaxation ( $\mu\text{SR}$ ) studies of polycrystalline  $\text{TbNi}_2\text{B}_2\text{C}$  [9] confirmed the presence of a small ferromagnetic component below about 8 K previously observed via neutron diffraction [7] and magnetization measurements [10, 11].

In addition to magnetic effects, strong phonon softening has been observed in superconducting  $R\text{Ni}_2\text{B}_2\text{C}$  single crystals with  $R = \text{Lu}, \text{Y}, \text{Er},$  and  $\text{Ho}$  [12–15], while no significant temperature dependence of the phonon spectra was detected for the nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  [16]. The superconducting transition temperature  $T_c$  systematically decreases for  $R\text{Ni}_2\text{B}_2\text{C}$  ( $R = \text{Lu}, \text{Y}, \text{Tm}, \text{Er},$  and  $\text{Ho}$ ) upon going from Lu ( $T_c = 16.6$  K) to Ho ( $T_c = 7.5$  K). This observation was interpreted by H. Eisaki *et al.* in terms of increasing coupling between the rare-earth magnetic moments and the conduction electrons [1], which suppressed superconductivity. For  $\text{TbNi}_2\text{B}_2\text{C}$ , this pair breaking could be strong enough to completely destroy superconductivity. An alternative possibility for the absence of superconductivity in this system is that electron-phonon coupling is weaker than in the superconducting compounds [16].

We investigated the strength of electron-phonon cou-

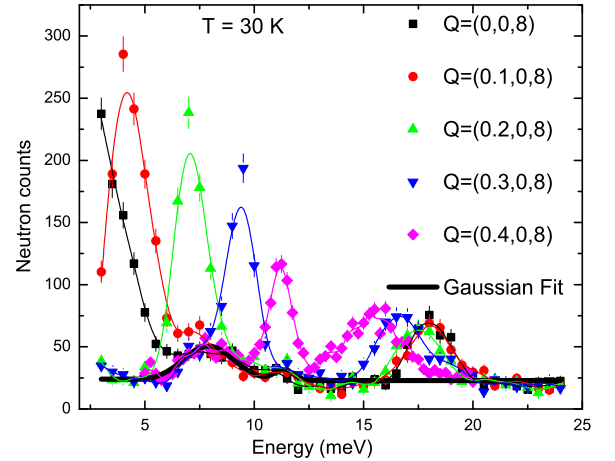


FIG. 1: (Color online) The  $\xi = 0$ ,  $\xi = 0.1$ ,  $\xi = 0.2$ ,  $\xi = 0.3$ , and  $\xi = 0.4$  spectra at  $T = 30$  K. The bold solid line represents the crystal electric-field excitations fit with a Gaussian line shape. Solid lines represent smoothed data.

pling in  $\text{TbNi}_2\text{B}_2\text{C}$  by detailed measurements of the temperature dependence of low-energy phonons in the magnetic nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  by inelastic neutron scattering. The observed softening of these phonon branches in this compound upon cooling from 300 to 30 K indicates that electron-phonon interactions in magnetic nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  are strong and, although weaker, are of the same order of magnitude as those in superconducting  $R\text{Ni}_2\text{B}_2\text{C}$  compounds.

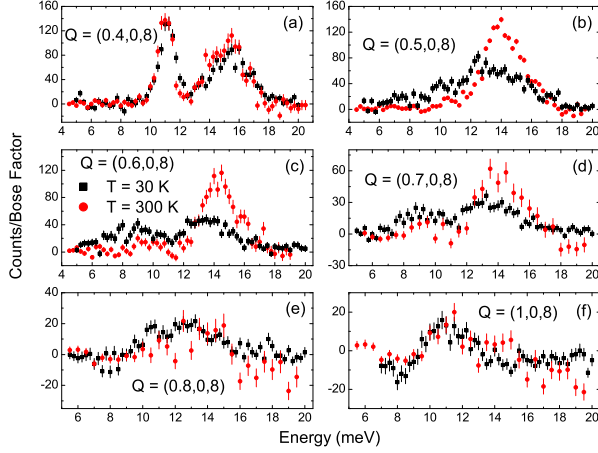


FIG. 2: (Color online) Background subtracted phonon spectra divided by the Bose factor taken at  $\xi = 0.4$  (a),  $\xi = 0.5$  (b),  $\xi = 0.6$  (c),  $\xi = 0.7$  (d),  $\xi = 0.8$  (e), and  $\xi = 1$  (f) at  $T = 30$  K (black squares) and  $300$  K (red circles).

## II. EXPERIMENTAL DETAILS

Intermetallic  $\text{RNi}_2\text{B}_2\text{C}$  borocarbides crystallize in the body-centered tetragonal  $\text{LuNi}_2\text{B}_2\text{C}$  structure with the space group  $I4/mmm$ , which consists of  $R\text{--C}$  layers separated by  $\text{Ni}_2\text{B}_2$  sheets [17]. All crystallographic parameters, i.e., the lattice constants  $a$  and  $c$  as well as the  $z$  position of boron, scale roughly with the ionic radius of the rare earth.  $\text{TbNi}_2\text{B}_2\text{C}$  compounds exhibit antiferromagnetic ordering at low temperatures below  $15$  K with the magnetic  $\text{Tb}^{3+}$  moments aligned along the  $a$  axis and modulated with the propagation vector  $\tau = (0.545, 0, 0)$  [7, 18]. No superconductivity has been detected in this compound down to  $7$  mK [19].

Rodlike  $\text{TbNi}_2\text{B}_2\text{C}$  single crystals were grown by the floating zone method using  $^{11}\text{B}$  isotope to avoid strong neutron absorption [20]. Specimens with a length of  $6$  mm were cut from the rods with  $6$  mm diameter. For the inelastic neutron-scattering experiments the samples were oriented in the  $(\mathbf{a} - \mathbf{c})$  scattering plane. The measurements were performed on the triple-axis spectrometer 1T1 at the Laboratoire Léon Brillouin, Saclay. Phonons propagating in  $[\xi \ 0 \ 0]$  direction, where  $\xi$  is a reduced wave vector, were recorded in the Brillouin zone centered at  $(0 \ 0 \ 8)$  by energy scans using a fixed final energy  $E_f = 14.8$  meV at temperatures between  $T = 2$  and  $300$  K.

## III. EXPERIMENTAL RESULTS

When phonons weakly couple to the conduction electrons their energies and lifetimes slightly harden and narrow upon cooling due to decreased anharmonicity. However, when phonons strongly couple to conduction electrons in the nearly nested regions of the Fermi surface, they soften and broaden upon cooling as the Fermi sur-

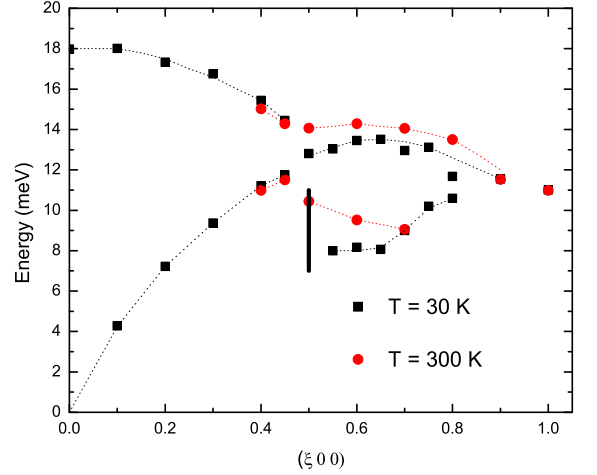


FIG. 3: (Color online) Low-energy phonons along  $[\xi \ 0 \ 0]$  at  $30$  and  $300$  K. The dashed lines are guides for the eye. The vertical black bar at  $\xi = 0.5$  denotes a broad phonon peak at  $30$  K containing also contributions from the next upper phonon branch.

face sharpens. Inelastic neutron- or x-ray-scattering measurements of phonons are the most direct way to identify the phonon modes strongly coupled to electrons. Due to the relatively large size of the available single crystal, we carried out our investigation using inelastic neutron scattering

Figure 1 illustrates how we determined the background that was subtracted from the raw data to obtain the phonon spectra. It shows energy scans near the zone center at  $T = 30$  K. The strongest peaks result from upward-dispersing acoustic phonons. The inelastic peaks between  $15$  and  $18$  meV originate from downward-dispersing optical phonons. In addition to phonons, crystal-electric-field (CEF) transitions are present between  $5$  and  $13$  meV [21]. For the purpose of studying the phonons, the CEF excitations contribute to the background. CEF excitations are dispersionless, and their form factor is nearly  $Q$ -independent in the narrow  $Q$ -range of interest. We determined them from the spectra where they do not overlap with phonons. The bold solid line in Fig. 1 represents the background, which includes Gaussian peaks due to CEF excitations and a straight line due to other energy-independent sources of background. It was subtracted from the data to isolate the one-phonon scattering shown in subsequent figures.

Figure 2 shows the temperature dependencies of phonon spectra at different wave-vector transfers ( $Q$ ) to the neutron at  $30$  and  $300$  K. The CEF contribution was subtracted from all low-temperature spectra as described above. We did not attempt to identify the contribution of the CEF excitations at  $300$  K, because they become weaker at high  $T$ , whereas phonons become stronger due to the Bose factor,  $n = 1/(1 - e^{-\hbar\omega/k_B T})$ . Thus their contribution to the scattering intensity becomes negligible. The data were divided by the Bose factor to cor-

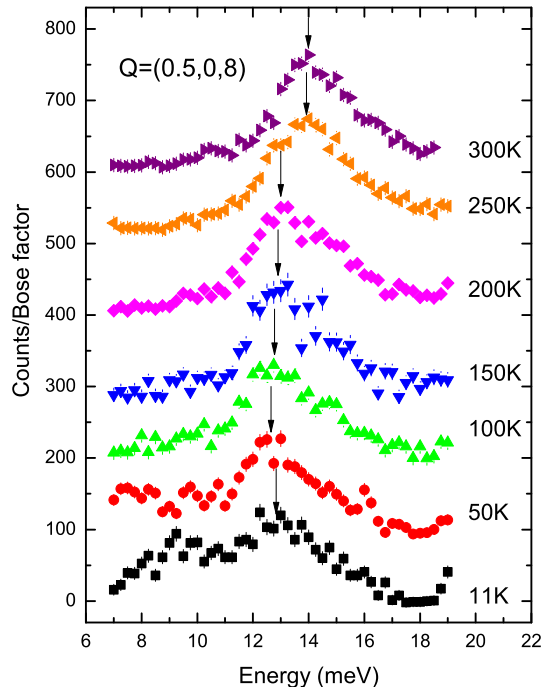


FIG. 4: (Color online) Phonon spectra divided by the Bose occupation factor for a wave vector  $(0.5, 0, 8)$  in the temperature range from 11 to 300 K. The curves are vertically shifted for clarity. The phonon-peak position is indicated by arrows.

rect for the temperature dependence of the one-phonon scattering, which also removed most temperature dependence from the background. High-temperature curves were scaled with the low-temperature ones by subtracting a constant, which is reasonable, because the background increases with  $T$ . Phonon spectra of the optical mode between 10 and 15 meV soften and broaden from 300 to 30 K at  $Q = (0.5, 0, 8)$ ,  $Q = (0.6, 0, 8)$ ,  $Q = (0.7, 0, 8)$ , and  $Q = (0.8, 0, 8)$ , whereas the spectra at  $Q = (0.4, 0, 8)$ ,  $Q = (0.9, 0, 8)$  (not shown), and  $Q = (1, 0, 8)$  are nearly temperature independent.

Figure 3 shows the dispersion of the two low-energy acoustic and optical phonon branches propagating along the  $[\xi 0 0]$  direction at  $T = 30$  and 300 K. The energy scans were fitted by Gaussian peaks. The dispersion of the two interesting phonon branches at  $T = 2$  K is similar to the dispersion measured in the temperature range  $T = 11 - 100$  K and there is no softening below 100 K [16]. However, strong softening of both branches appears between 100 and 300 K between  $Q = (0.5, 0, 8)$  and  $(0.8, 0, 8)$ . Our low-temperature data reveal a discrepancy in the energy range  $\hbar\omega = 8 - 10.5$  meV as compared to Ref. [16] which can be attributed to a different interpretation of CEF contribution. Unlike Ref. [16], we observe a strong dip of the low-energy phonon branch for wave vectors between  $Q = (0.5, 0, 8)$  and  $(0.75, 0, 8)$  at low temperatures, which is weaker but of the same or-

der of magnitude as the one found in superconducting  $\text{HoNi}_2\text{B}_2\text{C}$ . This result indicates that electron-phonon coupling in  $\text{TbNi}_2\text{B}_2\text{C}$  is strong enough to mediate superconductivity (perhaps with a lower  $T_c$ ) in the absence of a pair-breaking mechanism.

Figure 4 illustrates the temperature dependence of the optic phonon at  $Q = (0.5, 0, 8)$ . Here we show the raw data divided by the Bose factor without subtracting the background. The curves are vertically shifted with the arrows indicating the phonon peak position. The lowest temperature scan contains an additional feature at 9 meV to which both the acoustic phonon and the CEF excitation contribute. We have not measured the temperature dependence of the CEF excitation, so it is not possible to determine the temperature dependence of the acoustic phonon based on the available data. However, the optic phonon does not overlap with the CEF excitation and the data on its  $T$  dependence are unambiguous. We observe a softening from room temperature to  $T \cong 100$  K of 1 meV. Below  $\approx 100$  K, it shows no peak position shift.

#### IV. DISCUSSION AND CONCLUSIONS

Strong softening of two low-energy phonon branches was observed by neutron scattering in superconducting  $R\text{Ni}_2\text{B}_2\text{C}$  single crystals with  $R = \text{Lu}, \text{Y}$ , and  $\text{Er}$  [12–15]. Point-contact spectroscopy revealed strong electron-phonon interaction in the superconducting  $R\text{Ni}_2\text{B}_2\text{C}$  compounds with  $R = \text{Y}$  and  $\text{Ho}$ , in contrast to the nonsuperconducting  $\text{LaNi}_2\text{B}_2\text{C}$  [22], which suggests that the presence of superconductivity is controlled by the strength of electron-phonon coupling as opposed to magnetic pair breaking. However, our inelastic neutron scattering experiments on nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  clearly demonstrate strong electron-phonon coupling in this compound. Thus we conclude that superconductivity in  $\text{TbNi}_2\text{B}_2\text{C}$  is absent due to magnetic pair breaking.

Previous studies of the competition between magnetism and superconductivity in  $R\text{Ni}_2\text{B}_2\text{C}$  superconductors with the magnetic rare-earth elements  $R = \text{Lu}, \text{Tm}$ , and  $\text{Er}$  revealed a very weak coupling between the rare-earth magnetic moments and the conduction electrons due to a small conduction-electron density at the rare-earth site [1]. In contrast, a strong magnetic pair-breaking effect has been observed in  $\text{Dy}$  and  $\text{Tb}$  samples [1] which gives additional evidence that superconductivity in magnetic  $\text{TbNi}_2\text{B}_2\text{C}$  is indeed destroyed by the substantially strong interaction between the local magnetic moments and the conduction electrons. In summary, electron-phonon coupling in magnetic nonsuperconducting  $\text{TbNi}_2\text{B}_2\text{C}$  is strong enough to mediate superconductivity. This implies that its absence can only result from magnetic pair breaking.

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